

# Electric field induced coherent coupling of the exciton states in a single quantum dot

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The signature of coherent coupling between two quantum states is an anti-crossing in their energies as one is swept through the other. In single semiconductor quantum dots containing an electron-hole pair the eigenstates form a two-level system which can be used to demonstrate quantum effects in the solid state, but in all previous work these states were independent [1–5]. Here we describe a technique to control the energetic splitting of these states using a vertical electric field, facilitating the observation of coherent coupling between them. Near the minimum splitting the eigenstates rotate in the plane of the sample, being orientated at  $45^\circ$  when the splitting is smallest. Using this system we show direct control over the exciton states in one quantum dot, leading to the generation of entangled photon pairs.

It is well known that the exchange interaction in single semiconductor dots results in the exciton eigenstates being linearly polarised with an energy difference known as the fine-structure splitting (FSS,  $|s|$ ) [6, 7]. The magnitude of the FSS is determined by anisotropy in the strain, shape and composition of the dot, in addition to a contribution from the crystal inversion asymmetry [8]. When the FSS is smaller than the linewidth, the biexciton-to-exciton-to-empty cascade can lead to the emission of polarisation-entangled photon-pairs [9, 10], which has motivated much work on this subject.

Recently, theoretical work has suggested that for realistic strain-tuned dots a minimum in the FSS ( $s_0$ ) will be observed of the order of  $3 \mu\text{eV}$ , due to the symmetry of the crystal [11], but this has yet to be confirmed by experiment. Some success at tuning the FSS has been reported using strain [12] but this did not reach zero. Other tuning techniques such as magnetic field [1, 6], strong coherent lasers [2, 3], lateral electric field [4, 5, 13] and vertical electric field [14, 15] have been investigated but those that have been able to minimise the FSS have seen the states cross [1–5].

Arguably tuning the FSS with vertical electric field is the most practical technique yet reported, but the low confinement energies have limited the fields that can be applied to a few tens of  $\text{kVcm}^{-1}$  before carriers tunnel from the dot, so relatively small changes in FSS were observed [14, 15]. Here we demonstrate a design of heterostructure (Figure 1a) that allows much larger electric fields to be applied. Then both eigenstates of the exciton Stark shift at different rates, leading to a linear change in the FSS of over  $100 \mu\text{eV}$ . At appropriate fields we observed for the first time anti-crossings in the energies of the two exciton levels. Near

the anti-crossing hybridisation of the polarisation states of the upper and lower branches leads to their rotation in the plane of the sample. In a dot that has a small anti-crossing (below the homogeneous linewidths) we are thus able to demonstrate control of entangled photon pair emission using electric field. Other dots exhibit a large minimum splitting (FSS greater than the homogeneous linewidths) and a coherent superposition of the usual exciton states is observed.

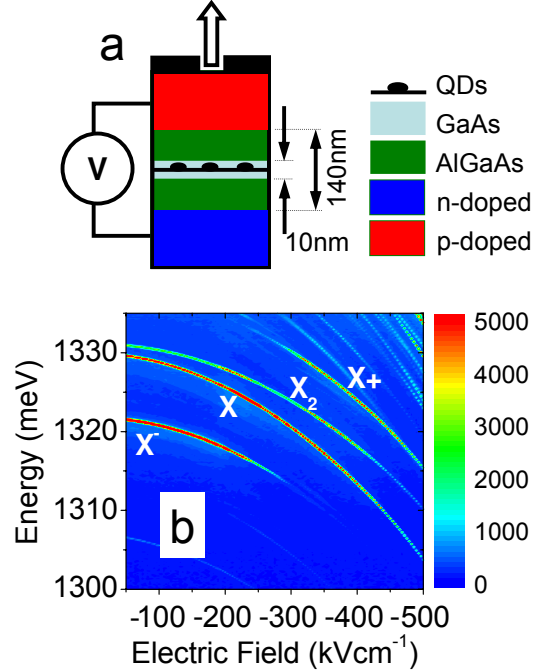


FIG. 1: Device design and observed giant Stark-shift of the excitonic transitions. **a** heterostructure design. **b** typical plot of photoluminescence versus electric field for a single quantum dot.

As electric field is applied all transitions Stark shift with the applied field (Fig. 1b), following the form  $E = E_0 + pF + \beta F^2$ , where  $p$  is the permanent dipole moment in the z-direction,  $\beta$  the polarisability and  $F$  the applied field [13, 16, 17]. At large values of FSS the exciton eigenstates can be thought of as radiating dipoles aligned along the  $[110]$  and  $[1\bar{1}0]$  crystal axes (see supplementary information), whose orientation is mapped onto the emitted photon's polarisation ( $X_{H/V}$  denoting photons of horizontal or vertical polarisation). Remarkably, away from zero FSS all neutral states display a linear change in the magnitude of the FSS with electric field that has gradient  $\gamma = -0.285 \pm 0.019 \mu\text{eV kV}^{-1}\text{cm}$  (Figure 2). Even the unusual dot we have identified that has its lowest energy exciton eigenstate orthogonal to all others in this sample, and thus is plotted on Fig. 2 with a negative  $s$ ,

has the same gradient. This value of  $\gamma$  is independent of the FSS at zero field, emission energy, binding energy and the Stark shift parameters  $p$  and  $\beta$ . The observed linear shift may be explained by the fact that the two eigenstates of the neutral exciton have slightly different confinement potentials in the two directions. This leads to different permanent dipole moments along the z-direction [17] giving  $p_H - p_V = \gamma$ . However, the polarisability of these states is unaffected by this in-plane anisotropy [16] and is controlled only by the height of the confinement potential, which is the same for both  $X_H$  and  $X_V$ . Thus the measured value of  $\gamma$  means the dipole moments of the two exciton eigenstates must differ by a few percent. The fact that  $\gamma$  is so similar in our ensemble suggests the dots all have comparable in-plane anisotropy, and that this value could be manipulated by changing the shape of the dots.

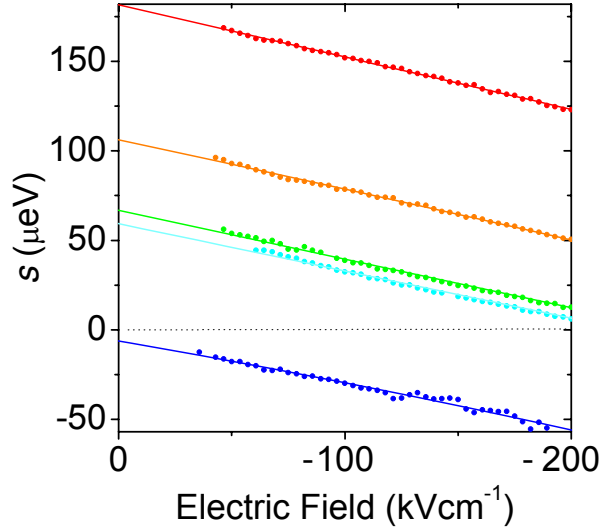


FIG. 2: Fine-structure splitting  $s$  as a function of electric field for 5 dots with naturally different  $s$  at low field, showing  $s$  varies in the same manner for all dots in this sample. One dot has an inverted fine-structure at all fields, with the lowest energy exciton state orthogonal to other dots we have studied, and is plotted with a negative  $s$ .

This ability to continuously tune the FSS over such a large range allows us to observe an anti-crossing in the neutral excitonic levels. The variation of the FSS with field is shown in Figure 3c for three dots, clearly indicating coherent coupling between these states. The splitting is well described by a simple model:

$$E \begin{pmatrix} \cos\theta \\ \sin\theta \end{pmatrix} = \begin{pmatrix} E_0 & s_0/2 \\ s_0/2 & E_0 - \gamma(F - F_0) \end{pmatrix} \begin{pmatrix} \cos\theta \\ \sin\theta \end{pmatrix}$$

where  $s_0/2$  quantifies the coupling between the states,  $F$  is electric field,  $F_0$  the field at minimal splitting and the energy levels move together with a rate  $\gamma$  in the absence of the coupling term  $s_0/2$ . This is the same form of equation that is used to describe coupled harmonic-oscillators [18], strong light-matter coupling [19], and anti-crossings in the states of molecular systems [20]. When  $\gamma(F - F_0) \gg s_0$  the natural basis to choose is that aligned with the crystal axes, and nearer  $(F - F_0) = 0$  the eigenstates will be a coherent mixture, with components  $\sin\theta$  and  $\cos\theta$ . In this system the parameter  $\theta$  is a real angle that describes the orientation of the eigenstates relative to the crystal lattice (Figure 3b). The eigenvalues,  $E_{\pm}$ , and eigenvectors are well known, with

$$E_{\pm} = E_0 - \frac{\gamma(F-F_0)}{2} \pm \frac{1}{2}\sqrt{\gamma^2(F-F_0)^2 + s_0^2}$$

$$\theta = \pm \tan^{-1} \left[ \frac{s_0}{\gamma(F-F_0) \pm (E_- - E_+)} \right]$$

We note that this simple model has degenerate solutions where the eigenstates rotate either clockwise or anti-clockwise when they approach  $(F - F_0) = 0$ . However, in practice we observe each dot has a clear preference to rotate one way or another in the plane of the sample, but never into the circular basis. Our measurements are consistent with the ensemble having no preferred direction of rotation. The origin of this handedness in individual dots is unknown, but may be determined by local defects or fields in the semiconductor.

Experimental data for three dots with varying sizes of anti-crossing and  $F_0$  are shown in Figure 3c and d, where all three dots rotate in the same direction. This data shows excellent agreement between the simple model and the experiment, indicative of a coherent coupling that can be activated with electric field.

A study of 22 dots revealed values of  $s_0$  in the range 0.7 to 42.9  $\mu\text{eV}$ , with lower values being observed more frequently (see supplementary information). No trend was observed between the magnitude of this value and other parameters associated with the electronic states of the dot, such as the FSS at  $F = 0$ ,  $F_0$ , the emission energy or  $(X(\phi) - X_2(\phi))$ .

Coupled pairs of oscillators have two normal mode frequencies whose values depend in part on the damping, or decoherence, affecting those individual oscillators [18]. For this

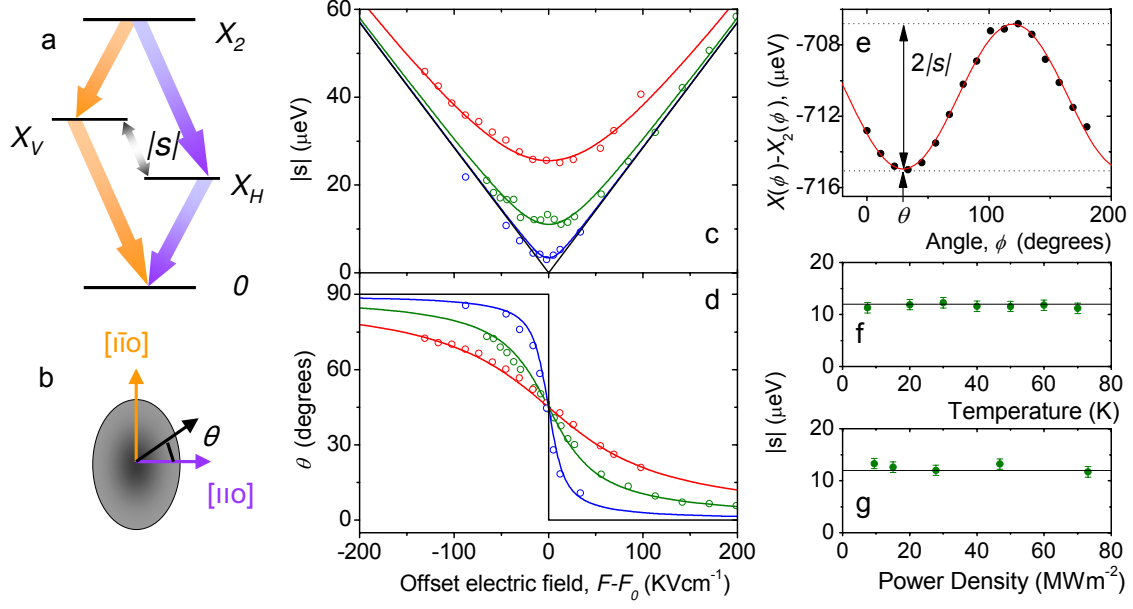


FIG. 3: Characteristics of electric-field-induced coherent coupling of exciton states in a single quantum dot. **a** Energy levels in the neutral cascade of a single dot. **b** Orientation of the eigenstates relative to the crystal axes. **c** and **d** plot  $|s|$  and  $\theta$  as a function of the electric field (offset by  $F_0$ ) for three quantum dots with values of  $s_0$  equal to 25.5, 12.0 and 3.0  $\mu\text{eV}$  and  $F_0$  of -140.5, -234.5 and -98.0  $\text{kVcm}^{-1}$ . The black line shows what would be expected from a dot with vanishing magnitude of anti-crossing. **e** The result of a typical measurement of the  $[X(\phi) - X_2(\phi)]$  from which we can extract  $|s|$  and  $\theta$ . **f** and **g** measurements of the amplitude of the anti-crossing for one of the dots as a function of temperature and excitation intensity, respectively.

reason we have investigated varying the sample temperature (from 4K to 70K, Figure 3f) and the excitation power, changing the ratio of exciton to biexciton intensity from 10:1 to 1.3:1, (Figure 3g) for the dot with  $|s_0|=12.0 \mu\text{eV}$ . Both of these factors will vary the decoherence experienced by a single state [21], but have no effect on the magnitude of the coupling. This suggests that although these external factors may cause decoherence of the individual states (such as the  $T_2$  time) it does not affect the time-scale on which the two-states dephase relative to each other [22].

We now show that we are able to generate entangled photons from a dot with a FSS of over 50  $\mu\text{eV}$  at zero field, by simply applying a voltage to the sample. The dot we study has an anti-crossing of amplitude  $s_0 = 1.5 \mu\text{eV}$  at a field of -240  $\text{kVcm}^{-1}$ . This splitting is below that required to observe entangled photon emission from the cascade [9, 10, 22]. To

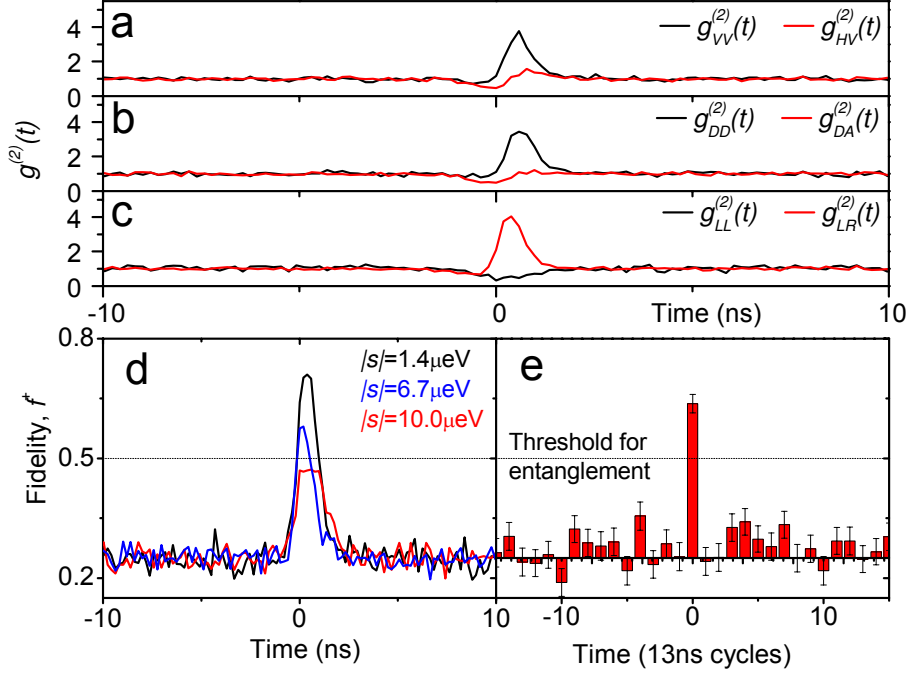


FIG. 4: Entanglement in the cascade emission of a dot with  $s_0 = 1.5 \mu\text{eV}$ . Polarised cross-correlations between  $X$  and  $X_2$  photons in three orthogonal polarisation bases: **a** rectilinear  $\{H, V\}$ , **b** diagonal  $\{D, A\}$  and **c** circular  $\{L, R\}$ . **d** Fidelity of the emitted state, for three different values of  $|s|$ . **e** Fidelity of the emission with pulsed optical excitation, considering only those X photons emitted within 100ps of a preceeding biexciton photon.

confirm this, polarised cross-correlation measurements were made between the  $X$  and  $X_2$  transitions at  $|s| = s_0$  (Figure 4a-c). The shapes of the peaks are dominated by the gaussian-like instrument response function with width  $\sim 600$  ps. As expected, strong correlation is observed in the rectilinear and diagonal bases (Figure 4a and b), and strong anti-correlation in the circular basis (Figure 4c), when the two photons are emitted at closely spaced times. The fidelity of the emission to the Bell state  $\Psi^+ = [|X^H X_2^H\rangle + |X^V X_2^V\rangle]/\sqrt{2}$ ,  $f^+$ , is given by  $[C_{\{H,V\}} + C_{\{D,A\}} - C_{\{L,R\}} + 1]/4$  [22] where  $C_{\{H,V\}}$  denotes the degree of polarisation correlation in the  $\{H, V\}$  basis. We obtain  $f^+ = 71 \pm 3\%$ . Similar measurements with pulsed excitation give  $f^+ = 64 \pm 3\%$  when averaging over all photons emitted within 100 ps of each other (Figure 4e). At this finite splitting the basis state that has maximum  $f^+$  rotates at a rate proportional to  $|s_0|$ , which combined with re-excitation causes  $f^+$  to fall to the classical value of 0.25 at times away from zero [23]. In both pulsed and CW measurements at  $s_0$ ,  $f^+$  is above the threshold of 0.5 confirming the emission of entangled

photon pairs has not been degraded by the application of such a large electric field. At different values of electric field and increased  $|s|$  CW correlation measurements confirm that as spectral distinguishability is introduced into the cascade  $f^+$  falls as expected (Figure 4d).

We have demonstrated an effective and versatile technique to control the FSS of a single quantum dot, which has enabled us to observe anti-crossings in the fine-structure of single dots. Such electric-field control is well suited to the incorporation of high-quality, low volume cavities facilitating higher efficiencies and cavity QED effects [24, 25]. In future, this technique will allow control of the fine-structure splitting and eigenstates of the exciton on a time scale faster than the radiative lifetime [26, 27], enabling manipulation of superpositions stored in these states.

## I. METHODS

The InAs dots are grown at the centre of a 10nm GaAs quantum well clad with a short period superlattice equivalent to  $\text{Al}_{0.75}\text{Ga}_{0.25}\text{As}$ . Doping that extends into the superlattice allows application of an electric field along the growth direction. This  $p-i-n$  device has a  $d = 140\text{nm}$  thick  $i$ -region and is encased in a weak planar microcavity consisting of 14/4 periods below/above the dot-layer. Thus the applied electric field is calculated to be  $(V - V_{bi})/d$  where the built-in potential,  $V_{bi}$ , is 2.2V. The  $\text{Al}_{0.75}\text{Ga}_{0.25}\text{As}$  barrier on either side of the quantum well controls the charging of the dot. Although the dot layer is positioned an equal distance from the n and p contact the tunneling rates will differ substantially due to the lower effective mass of the electron and different confinement energies of the carriers.

Excitation and photon collection occurs through an opaque metallic film on the sample surface, patterned with micron-diameter apertures (Figure 1a). During spectroscopy the samples were excited by a CW 850 nm laser diode, which creates carriers in the wetting layer and dot only. For cross-correlation measurements a Ti-Sapphire laser operating in either pulsed or CW regime excited the dots at 850 nm.

Single dots have a characteristic spectral arrangement of optical transitions that can readily be identified as exciton ( $X$ ), biexciton ( $X_2$ ) and charged excitons ( $X^+, X^-$ ), where this notation refers to the initial state. For neutral  $X_2$ -to- $X$ -to-empty cascade we are able to induce Stark shifts of 25 meV at  $500\text{ kVcm}^{-1}$ . Study of several dozen dots emitting in the range 1310-1340 meV showed the FSS at  $50\text{ kVcm}^{-1}$  displayed a Gaussian distribution



centred on  $109\text{ }\mu\text{eV}$  with width  $67\text{ }\mu\text{eV}$ . Such a distribution fits well with the trend previously reported for dots in GaAs[28]. These observations confirm that placing the dots in a quantum well has not changed their electronic properties.

We determine  $|s| = |E_+ - E_-|$  and  $\theta$  by measuring spectra polarised at multiple angles  $\phi$  to the [110] crystal direction, extracting the energy difference between the exciton and biexciton transitions,  $(X(\phi) - X_2(\phi))$ . This technique eliminates small energy shifts induced by rotation of the polarisation optics giving measurements of  $s(\phi)$  with sub- $\mu\text{eV}$  accuracy [1, 28]. When  $|s|$  is below the resolution of the system used for this measurement ( $\sim 40\text{ }\mu\text{eV}$ ) we observe a sinusoidal variation in  $(X(\phi) - X_2(\phi))$  (Figure 3e) from which we determine the orientation angle,  $\theta$  and magnitude of the FSS,  $|s|$ , at each field.

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## III. AUTHOR CONTRIBUTIONS

The samples were grown by I.F, C.A.N, D.A.R and processed by R.B.P. The optical measurements were made by A.J.B, M.P and R.M.S. A.J.S. guided the work. All authors discussed the results and their interpretation. A.J.B wrote the manuscript, with contributions from the other authors.

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